

REMARKS

At the outset, applicant wishes to thank Examiner Jonas N. Strickland for the courtesies extended during the personal interview held on October 6, 2004 with the undersigned attorney. The Examiner's careful attention to the application on that occasion is sincerely appreciated.

Claims 9-26 were rejected under 35 USC §102(b) as being anticipated by LI et al. 5,149,512. The Official Action states that the patented reference discloses a catalytic reduction process for the reduction of nitrogen oxides using methane in the presence of oxygen. The methane is stated to serve as a reducing agent, and the catalyst is allegedly a promoted iron-containing zeolite. The precious metal may be rhodium, and the term NO_x is stated to include nitrous oxide in column 3, lines 56-58. The methane to NO_x ratio ranges from 0.2 to 10, and the process is carried out at a temperature of about 250°C and pressures between 1 and 300 atmospheres. It is concluded that since LI et al. disclose the same process, it would have been inherent and anticipated to achieve the emission of carbon monoxide and of the hydrocarbon being less than 100 ppmv.

Reconsideration of the above rejection is respectfully requested for the following reasons.

As was explained during the interview, in order to consider the relevance of the LI et al. reference towards the present application, it is essential to understand what LI et al.

are disclosing and what the present application is claiming, respectively. Initially, the teachings of the LI et al. reference will be discussed, followed by the sufficiency of such reference. During such discussions, the novelty and non-obviousness of the present invention will be assessed.

1. TEACHING OF LI et al.

The background of LI et al. teaches that NO_x refers to nitrogen dioxide NO₂ and nitric oxide NO (column 1, lines 16-18) and that the prior art does not disclose a catalytic process for destroying NO_x in oxygen rich combustion products (column 1, lines 31- column 3, line 36).

The brief summary, detailed description and claims of the patented reference teach that zeolites with Si/Al ratio greater than or equal to 2.5, which are exchanged with a cation (singular: the reference does not *explicitly* disclose that the zeolite can be exchanged with *combinations of these cations*) and wherein the cation is selected from Co, Ni, Fe, Cr, Rh and Mn can be used for the conversion of NO_x with CH₄ (column 3, lines 45-55). It is further taught that the term NO_x comprises amongst others N₂O.

Further, the detailed description teaches two alternate embodiments.

In the first embodiment, the metal exchanged zeolite catalyst is subjected to a further metal exchange with cations,

with metals represented by the third period transition metals and members of groups 8, 9 and 10 (see column 3, line 63 - column 4, line 2; column 7, lines 15-28; claim). It is unclear whether the metals for the further metal exchange have to be members of *both* the third period transition metals and of group 8, 9 and 10, but the examples of preferred cations in column 7, lines 23-24 *may suggest* that this is not the case. Hence, this embodiment may comprise zeolites which are exchanged with Co, Ni, Fe, Cr, Rh or Mn and which are *further exchanged* with Fe, Co, Ni, Ru, Pd, Ag, Hf, Ta, W, Re, Os, Ir, Pt, Au or Hg. Note however that as (preferred) cations, only **Co, Ni, Fe, Mn** and **Ag** are *explicitly* disclosed by LI et al. as second elements. No further elements belonging to this group of third period transition metals and group 8, 9 and 10 than Co, Ni, Fe, Mn and Ag are *explicitly* disclosed.

In the second alternate embodiment, the ion exchanged zeolite is impregnated with various anionic and neutral species. Suitable impregnating moieties include oxidizing metals selected from groups 5, 6, 7 and 11 (column 4, lines 7-8; column 7, lines 29-54; claims 7-9). Hence, this embodiment comprises zeolites which are exchanged with Co, Ni, Fe, Cr, Rh or Mn and which are further impregnated with V, Cr, Mn, Cu, Nb, Mo, Tc, Ag, Ta, W, Re, Au. Note that as metals selected from groups 5, 6, 7 and 11 only **Ag**, and **oxides of Nb, Mo, V** and **Mn** are *explicitly* mentioned (column 7, lines 40-42; claim 9).

The examples in LI et al. show that most of the examples are performed with ZSM-5 (17 out of 21). Further, 11 examples (runs 2, 4, 7a, 8, 9, 10, 11, 11a, 11b, 13, 14) are directed to ZSM-5 exchanged with one type of metals, of which 3 are reference examples (runs 2, 7a and 13), and from these eight single exchanged zeolites, *only 2* are directed to Fe-exchanged zeolites (run 11b and 14). Note however that these Fe-exchanged zeolites are inferior to Co, Ni or Mn exchanged zeolites (table 2).

Further, from the examples, 9 examples (runs 3, 5, 6, 7, 17, 18, 19, 20, 21) are directed to ZSM-5 provided with 2 types of metals (of which 1 is a reference example (run 3)), and from these 8 examples of zeolites having two metal types, *none is directed to a zeolite with Fe*, but all 8 examples (100%) are directed to zeolites exchanged with cobalt. Co is apparently the preferred ion.

Summarizing the teaching of LI et al. and comments with respect to novelty of the present invention:

LI et al. and the examples therein disclose the conversion of NO (tables 1A, 2-9). This is stressed by the summary of the experiments in column 15, lines 24-33. A person having ordinary skill in the art knows that catalysts which are suitable for conversion of NO and NO₂ are not necessarily suitable for conversion of N₂O. This is clearly demonstrated in

the two accompanying 37 CFR 1.132 declarations of Dr. Rudolf Willem van den BRINK and Dr. Johannis Alouisius Zacharias PIETERSE.

Since the person skilled in the art will derive that the catalysts mentioned in LI et al. are only suitable for the reduction of NO, NO₂, etc. (i.e. NO_x) and not N₂O, it is respectfully submitted that claims 9 and 18 are novel over LI et al.

The patented reference discloses (a) that zeolites exchanged with Fe can be used for the conversion of NO with methane (table 2) and also discloses (b) in general that zeolites exchanged with Co, Ni, Fe, Cr, Rh or Mn can be used for the conversion of NO_x with CH₄.

Hence, the patented reference does not explicitly disclose combination of the herein-claimed elements of (1) using a Fe exchanged zeolite for the conversion of (2) N₂O with saturated hydrocarbons according to claims 9 and 18 of the present invention: thus new claims 9 and 18 are novel over LI et al.

Of all examples of zeolites containing two ions, none of them contain iron, and all of them contain cobalt as ion with which the zeolite is first exchanged. None of the 162 possible metal combinations include a combination with Fe.

Hence, the patented reference does not explicitly disclose the combination of the herein-claimed elements of (1)

using a *Fe exchanged zeolite* which is (2) further promoted with Ru, Rh, Pd or Au for (3) the conversion of N_2O with saturated hydrocarbons according to claims 9 and 18 of the present invention: thus new claims 9 and 18 are novel over LI et al.

In all the examples of zeolites of LI et al. containing two ions, at least Co as catalytic ion is present. However, *none of these Co-containing zeolites further comprise Ru, Rh, Pd or Au.*

Hence, the patented reference does not *explicitly* disclose the combination of the herein-claimed elements of (1) using a *Fe exchanged zeolite which is* (2) *further promoted with Ru, Rh, Pd or Au* for (3) the conversion of N_2O with saturated hydrocarbons according to claims 9 and 18 of the present invention: thus claims 9 and 18 are novel over LI et al.

The first and second alternate embodiments only explicitly disclose the possibility of zeolites being exchanged with Co, Ni, Fe, Cr, Rh or Mn further being provided with (embodiment 1) Co, Ni, Fe, Mn and Ag or with (embodiment 2) Ag, and oxides of Nb, Mo, V and Mn.

Hence, the LI et al. reference does not *explicitly* disclose the combination of the herein-claimed elements of using (1) a *zeolite containing the couples Fe and Ru, Fe and Rh, Fe and Pd or Fe and Au* for (2) the conversion of N_2O with saturated hydrocarbons according to claims 9 and 18 of the present invention: thus claims 9 and 18 are novel over LI et al.

2. SUFFICIENCY OF DISCLOSURE OF LI et al.

The disclosure of LI et al. with respect to zeolites comprising two metal ions for the use of reduction with methane of N_2O , is relevant in determining novelty of the present invention.

a. The two embodiments of LI et al.

Referring to the two embodiments, the first alternate embodiment implicitly discloses 90 possible combinations of couples of metal ions. At least 4 of these implicitly disclosed possible combinations make no sense (Fe-Fe, Co-Co, Mn-Mn and Ni-Ni). Further, some of the implicitly disclosed possible combinations probably make no sense from a chemical perspective (e.g. ion exchanging with W or Hg?). Note further that also Mn is given as preferred cation, which is neither a third period transition metal nor a group 8, 9 or 10 metal!

Hence, in view of all these unclarities, it is respectfully submitted that the patented reference fails to disclose at all how and with what kind of metal ions the metal exchanged zeolites can be exchanged in this first alternate embodiment.

Referring to the second alternate embodiment, 72 combinations of 2 metals are implicitly disclosed. At least 2 of these implicitly disclosed possible combinations make no sense (Cr-Cr and Mn-Mn). Further, some of these implicitly disclosed

possible combinations probably make no sense from a chemical perspective (e.g. Tc?). Next to that, it is in a chemist's view unclear why these metals are anionic or neutral species. This embodiment becomes even more obscure, since it stays undisclosed why Ag (explicitly disclosed; column 7 lines 24 and 40) Ta, W and Re are *anionic or neutral* species in alternate embodiment 2, but *cationic* species in alternate embodiment 1.

Further, the person having ordinary skill in the art is not taught how in this case these impregnations should be performed and how such metals must be impregnated as *anionic or neutral* (or perhaps both?) species. Note to this respect further that another example of this alternate embodiment, example 4, discloses that $\text{Nb}(\text{HC}_2\text{O}_4)$ is dissolved in $\text{H}_2\text{C}_2\text{O}_4$ and H_2O , thereby already showing that neither an anionic species, nor an neutral species, but a cationic species is impregnated on the zeolite (Nb is dissolved as cationic species, completely analogues to e.g. Co-acetate ((with Co as *cation* in solution) in e.g. example 1).

Hence, in view of all these unclarities, it is urged that the patented reference fails to disclose at all how, and with what kind of metal ions the metal exchanged zeolites can be impregnated in the second alternate embodiment.

**i. Summarizing the disclosure of the two embodiments
of LI et al. and comments with respect to novelty
of the present invention**

The patented reference fails to sufficiently disclose how the zeolites should be promoted by a second metal: the groups with second metals are not clearly defined, chemical unacceptable elements are (inherently) suggested, combinations of metals which make no sense are suggested, metals serving both cationic and anionic properties are suggested, inconsistency between description and examples (example 4) are presented.

It is by now well settled that when a claimed invention is not identically disclosed in a reference, and instead requires picking and choosing among a number of different options disclosed by the reference, then the reference does not anticipate. See *Mendenhall v. Askec Industries, Inc.*, 13 USPQ2d 1956 (Fed Cir. 1989).

b. The reduction of N₂O with methane

None of the examples disclose the conversion of N₂O (tables 1A, 2-9) but only show that NO can be converted. This is confirmed by the later publication of Li and Armor in Appl. Catal. B 3, 1993, on page 56), a copy of which is enclosed with each of the accompanying 37 CFR 1.132 declarations for the Examiner's convenience.

The patented reference itself has different definitions for NO_x (on the one hand *including* N₂O: column 3, lines 56-58 and column 4, lines 44-47; and on the other hand *without* N₂O, column 1, lines 16-18, column 15, lines 24-30), thereby guiding the person skilled in the art, to the interpretation of NO_x that is

most likely: i.e. the interpretation of NO_x not being N_2O but being NO (and NO_2 , etc.), i.e. the interpretation as can further be derived from column 10, lines 6-8 in conjunction with column 10, lines 35 and 61, the interpretation of tables 1a, 2, 3, 4, 5, 6, 7, 8, and the interpretation of the header of table 9 and the remarks under table 9, each showing that NO_x is to be understood as NO but not as N_2O .

Further, the person skilled in the art knows that the reduction of N_2O follows another mechanism than the reduction of NO and different catalysts are used. This is also conceivable, since the oxidation states of nitrogen in the compounds are different. Due to the different chemical behavior, the species are performing differently when brought into contact with a catalyst.

Proof of this can be found in Appl. Catal. 64, 1990, L1-L4 (copy furnished with each accompanying 37 CFR 1.132 declaration), where it appears that reduction of NO_x with propane over a zeolite catalyst leads to the formation of N_2O (see also table 1 of this publication), which shows that this catalyst does not reduce NO_x and N_2O , but only NO_x : this zeolite catalyst is therefore not suitable for the reduction of N_2O . Further proof can be found in Appl. Catal. B 25, 2000, 191-203 (copy provided with each accompanying 37 CFR 1.132 declaration), wherein it is shown that a first catalyst is used for the reduction of NO_x , whereby however large amounts of N_2O are formed. A second,

reduction of N_2O but performs as catalyst badly for the reduction of NO (Appendices 1 - 3). From these data it appears that there is a general trend that catalysts suitable for N_2O reduction are not well applicable for NO reduction in SCR processes (and vice versa).

i. Summarizing the disclosure of N_2O reduction with methane in LI et al. and comments with respect to novelty of the present invention

Hence, the person skilled in the art will understand from a fair reading of LI et al. that the catalysts described are suitable for the reduction of NO, NO_2 etc., but not for the reduction of N_2O . Accordingly, the patented reference is not an enabling disclosure for the reduction of N_2O with methane in the presence of a catalyst, hence, new claims 9 and 18 are novel over LI et al.

3. NONOBVIOUSNESS

The patented reference discloses that Fe exchanged ZSM-5 is clearly worse for the conversion of NO with methane. Even if the person skilled in the art would have considered using Fe-exchanged zeolites, he would have to make the following steps to come at the present invention: (a) choose Fe-exchanged zeolites (bad performance) instead of the clearly favorable Co-exchanged zeolites, (b) erroneously think that (Fe-)zeolites which are suitable for NO-conversion will always be suitable for the conversion of N_2O (not disclosed in the patent), (c) for some

zeolite based catalyst is necessary for the decomposition of N_2O . This shows that a zeolite based catalyst only cannot reduce efficiently both NO_x (NO) and N_2O .

In order to prove that what is stated here is known to a person having ordinary skill in the art, there is submitted herewith two separate 37 CFR 1.132 declarations, wherein the declarants have performed additional experiments, which indeed show that catalysts that are suitable for reduction of N_2O are not necessarily suitable for the reduction of NO. Applicant is convinced that a person skilled in the art reading the LI et al. patent will unambiguously derive that the catalysts mentioned in this document are only suitable for the reduction of NO, NO_2 , etc. (i.e. NO_x) and not N_2O .

The additional experiments are explained and presented in each 37 CFR 1.132 declaration as "Appendices Additional Experiments". The experiments clearly show that under exhaust gas conditions ZSM-5 exchanged with Fe (not according to the invention) provides good catalysts for the reduction of N_2O but performs badly for the reduction of NO with propane. The experiments further show that under exhaust gas conditions, ZSM-5-Fe,Pd (a catalyst according to the invention) is a good catalyst for the conversion with methane of N_2O , but a bad catalyst for the conversion with methane of NO. The experiments also show that under exhaust gas conditions yet a third catalyst, not according to the invention, provides a good catalyst for the

undisclosed reason decide to exchange or impregnate? in anionic or neutral form? (not from the teachings of the reference) with Ru, Rh, Pd or Au (which selection is also not disclosed). This combination of steps is highly improbable and not likely in view of the teachings of LI et al. (e.g. there is an incentive for using Co exchanged zeolites but no incentive at all to use Fe-exchanged zeolites, and there is no incentive to use Ru, Rh, Pd or Au as second metal).

Hence, it is respectfully submitted that the LI et al. reference neither discloses nor suggests a Fe exchanged zeolite, which is further provided with Ru, Rh, Pd or Au for the conversion of N_2O with saturated hydrocarbons. Therefore, claims 9 and 18 are unobvious with respect to LI et al.

4. The present invention

The present invention claims the use of zeolites which contain two catalytically active metals, of which one is at least Fe and the second metal is one selected from the group consisting of Ru, Rh, Pd and Au, for the reduction with saturated hydrocarbons of N_2O .

These zeolites are called promoted Fe-containing zeolites, since next to Fe, which may e.g. be applied to the zeolite by ion exchange, the zeolite further contains other active species selected from the group consisting of Ru, Rh, Pd and Au, which may e.g. be applied to the zeolite (containing Fe) by impregnation. The zeolite which is used in the process of the

In view of the present amendment, the accompanying 37 CFR 1.132 declarations, and the foregoing remarks, therefore, it is believed that this application has been placed in condition for allowance. Entry of the present amendment, reconsideration and allowance of claims 9-26 are accordingly earnestly solicited.

In the event that there are any questions relating to this amendment or to the application in general, it would be appreciated if the Examiner would telephone the undersigned attorney concerning such questions so that the prosecution of this application may be expedited.

The Commissioner is hereby authorized in this, concurrent, and future replies, to charge payment or credit any overpayment to Deposit Account No. 25-0120 for any additional fees required under 37 C.F.R. § 1.16 or under 37 C.F.R. § 1.17.

Respectfully submitted,

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present invention is therefore a zeolite that can be described by:

Ru-Fe-Zeolite (e.g. Ru-Fe-ZSM-5)

Rh-Fe-Zeolite (e.g. Rh-Fe-ZSM-5)

Pd-Fe-Zeolite (e.g. Pd-Fe-ZSM-5)

Au-Fe-Zeolite (e.g. Au-Fe-ZSM-5)

These zeolites are claimed for use in the method for the catalytic reduction of N_2O , which method provides a very suitable way to remove the environmental unfriendly N_2O from e.g. exhaust gases. This method can e.g. be used next to the method of LI et al., since these methods are complementary, the one removing N_2O , the other one removing NO_x (NO , NO_2 , etc.) The method of the invention provides additional benefits over prior art methods and may be of great value for basic chemicals production plants that emit N_2O , like those for the production of nitric acid or caprolactam, etc.

As is reflected in the Examiner's Interview Summary, independent claims 9 and 18 have been amended so as to clarify that the catalyst comprises a zeolite which contains at least one combination of metals selected from the group consisting of iron and ruthenium, iron and rhodium, iron and palladium and iron and gold, for the catalytic reduction of nitrous oxide. The Examiner kindly indicated that these changes would appear to place the application in condition for allowance.

APPENDIX:

The Appendix includes the following items:

- a 37 CFR 1.132 Declaration of Dr. Rudolf Willem van den BRINK
- a 37 CFR 1.132 Declaration of Dr. Johannis Alouisius Zacharias
PIETERSE